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Abstract

Developed for radionuclide standardization using liquid scintillation, the Triple to Double Coincidence Ratio (TDCR) method is applied using coincidence counting obtained with a specific three-photomultiplier system. For activity determination, a statistical model of light emission is classically used to establish a relation between the detection efficiency and the experimental *TDCR* value. At LNE-LNHB, a stochastic approach of the TDCR modeling was developed using the Monte Carlo code Geant4. The interest of this TDCR-Geant4 model is the possibility to simulate the propagation of optical photons from their creation in the scintillation vial to the production of photoelectrons in photomultipliers.

As an alternative to the use of radionuclide sources, first TDCR measurements are presented using a miniature x-ray tube closely coupled to the scintillation vial. The objective of this new set-up was to enable low-energy depositions (lower than 20 keV) in liquid scintillator in order to study the influence of both time and geometrical dependence between PMTs already observed with radioactive sources. As for the statistical TDCR model, the non-linearity of light emission is implemented in the TDCR-Geant4 model using the Birks formula which depends on the *kB* factor and the scintillation yield. Measurements performed with the x-ray tube are extended to the assessment of these parameters and they are tested afterwards in the TDCR-Geant4 model for activity measurements of ^{3}H.

Keywords: TDCR method, Radionuclide metrology, X-ray tube, Geant4 simulation

1. Introduction

The Triple to Double Coincidence Ratio (TDCR) method is widely applied in National Metrology Institutes for primary radionuclide standardization (Broda et al., 2007). Based on a specific liquid scintillation (LS) counter equipped with three photomultiplier tubes (PMTs), the activity is determined using the experimental TDCR ratio given by double and triple coincidences of emitted scintillation photons between PMTs. To this end, a statistical model of light emission is implemented according to several assumptions such as stochastic independence between PMTs (Bobin et al., 2012a), Poisson distribution of photoelectrons in PMTs, etc. In the statistical modeling, double and triple coincidences between PMTs are calculated using an analytical expression based on the probability to count at least one photoelectron as a result of an energy deposition. However, the statistical approach does not

account for the optical and geometrical properties of the detector in terms of refraction and reflection processes of optical photons.

At LNE-LNHB (Laboratoire National Henri Becquerel), an alternative to the statistical TDCR model is studied using the Geant4 simulation toolkit (Agostinelli et al., 2003) in order to simulate the transport of charged particles and optical photons resulting from scintillation and the Cherenkov effect. With the TDCR-Geant4 model, optical photons are simulated from their creation in the optical cavity to the generation of photoelectrons in PMTs leading to double and triple coincidences. From previous studies, several results were obtained from the application of the TDCR-Geant4 model in the case of low-energy depositions (lower than 20 keV). In particular, simulations carried out for mono-energetic depositions (5 keV, 8 keV and 12 keV) revealed the existence of a stochastic dependence between PMTs due to geometrical optics (or geometrical stochastic dependence). This effect results from the sensitivity of the photon distribution between PMTs to the location of light emission inside the scintillator volume combined with reflection and refraction processes occurring at the different interfaces of the optical cavity. As shown with the TDCR-Geant4 model for discrete-energy emitters, the consequence is an overestimation of detection efficiencies obtained with the standard statistical model. These simulation results were experimentally confirmed in the case of ^{51}Cr standardization (this radionuclide disintegrates by electron capture with maximum energies of x-ray photons and Auger electrons mainly comprised between 4 keV and 6 keV).

The influence of the geometrical stochastic dependence (i.e. due to geometrical optics) on detection efficiencies is directly observed by a shift of the counting rates for the same experimental *TDCR* value when using diffusive polyethylene vials instead of glass vials. This shift observed between the two types of vials leads to an increase of the activity calculation which was also mentioned by Simpson et al. (2010) in the case of ^{55}Fe measurements.

In the present paper, first TDCR measurements using a miniature x-ray tube are presented as an alternative to radionuclide sources. In order to obtain interactions of low-energy x-rays in the liquid scintillator, the end-window transmission-target x-ray tube (MAGNUM® 40 kV manufactured by Moxtek) was coupled to a scintillation vial. The first objective was to test the reproducibility with the x-ray tube of the two types of stochastic dependence identified with radioactive LS sources: 1) when the coincidence resolving time is too short with regard to the time distribution of photons between PMTs; 2) due to geometrical optics as previously discussed. These two types of stochastic dependence were first investigated experimentally for a low-energy deposition set to 2.7 keV. The influence of the liquid scintillator on the evolution of the counting rates according to the resolving time was checked. The modification due to geometrical optics of the relation between the detection efficiency of double coincidences and *TDCR* values was also tested using glass and polyethylene vials.

As for the statistical model, the Birks formula is implemented in the TDCR-Geant4 model to account for the non-linearity of light emission as a consequence of ionization quenching. The associated parameters [*kB* factor; scintillation yield] were assessed using measurements performed with the x-ray tube for 3 different x-ray energies (2.7 keV, 8.7 keV and 17.3 keV). These parameters are tested afterwards in the case of the standardization of ^{3}H with the TDCR-Geant4 model.

2. Experimental TDCR set-up with a miniature x-ray generator

As already described in previous studies (Thiam et al., 2010), the detection system is composed of three XP2020Q photomultipliers equipped with a fused silica window. Counting vials (glass or polyethylene) are hung inside a spherical cavity made of Teflon®. For coincidence counting, the electronics chain is composed of a fast amplifier (Phillips scientific model 777) and a Constant Fraction Discriminator (CFD) module (Canberra Quad CFD 454). The logical signals provided by these front-end electronics are used to feed either a MAC3 module (Bouchard and Cassette, 2000) that has been modified to set variable resolving times or an FPGA (Field Programmable Gates Array)-based digital system which is also used as a time-to-digital converter combined with the processing of counting losses according to the live-time technique using extendable dead-times (Bobin et al., 2012b).

The interest of integrating a miniature x-ray tube into the detection set-up is the ability to perform TDCR measurements with low-energy x-ray photons in the liquid scintillator as an alternative to radionuclide sources. As depicted in Fig. 1, this small-size x-ray generator (~55 mm length, ~29 mm diameter) is directly coupled to the counting vial (for the x-ray beam, a small hole is machined in the plastic cap). The x-ray tube is designed with a transmission-target end-window configuration with tungsten for the anode material. Three energies have been selected for this study: 2.7 keV, 8.7 keV and 17.3 keV. The x-ray-beam energy is tuned by applying an appropriate high-voltage potential and by using an attenuation film placed at the output of the x-ray tube. The upper-energy cutoff is defined by the K-shell absorption edge of the film component. The x-ray energy spectrum is sharpened using a suitable attenuation thickness (see Table 1 for the settings used for the 3 x-ray energies). For instance, the 2.7 keV-energy emission is obtained using the following settings: high voltage set to 3 kV, and PVC film of ~100 μm thickness (K-shell electron binding energy of chlorine equal to ~2.82 keV). The spectrum shapes at the end-window were measured with a Silicon Drift Detector (SDD, AXAS-M manufactured by KETEK) calibrated using ^{55}Fe and ^{241}Am sources.

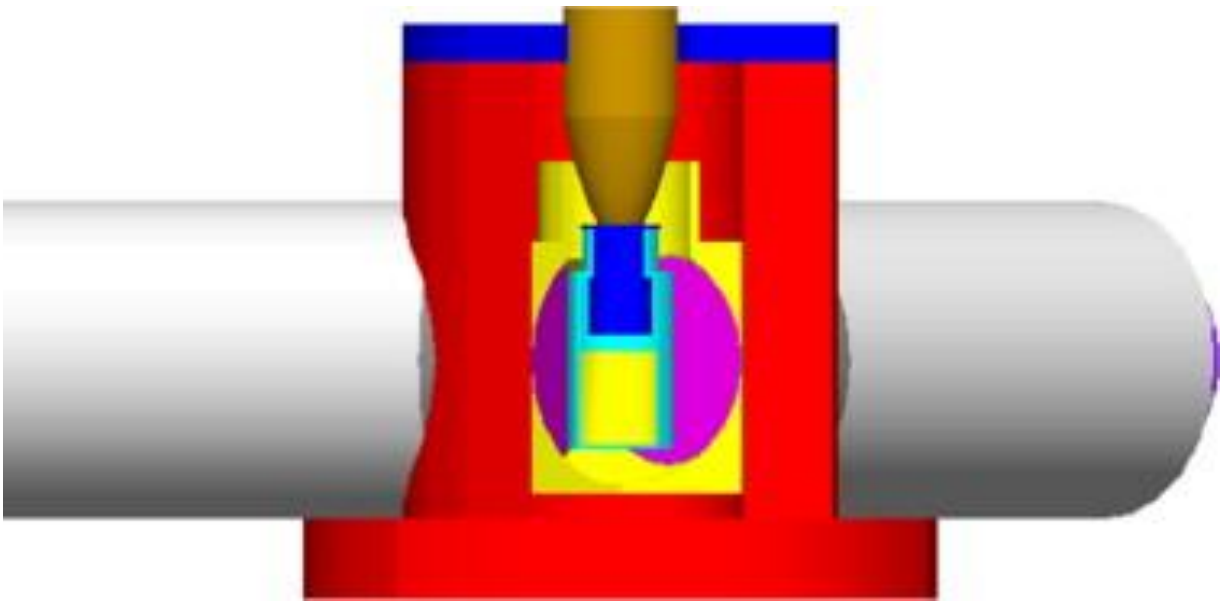

Fig. 1. Geometrical modeling with Geant4 of the LS-counter vial coupled with the miniature x-ray tube. The collimator (used for [*kB*; S] measurements) is also included inside the vial, which was hung between PMTs in the optical cavity.

X-ray energy (keV)	Attenuation film
2.7	Polyvinyl chloride (100 μm)—cutoff energy: 2.8 keV
8.7	Pure copper (300 μm)—cutoff energy: 9 keV
17.3	Pure zirconium (800 μm)—cutoff energy: 18 keV

Table 1. Attenuation films used with the x-ray tube set for several energies. The cut-off energy is defined by the K-shell electron binding energy of the film component.

3. Experimental results

The sensitivity of coincidence counting with varying resolving times has already been observed by several laboratories in the case of low-energy radionuclides (Steele et al., 2009, Bobin et al., 2010a). According to complementary investigations, this effect occurs when the coincidence resolving time is too short with regard to the arrival-time distribution of photoelectrons, leading to coincidences between PMTs. As observed for the standardization of ^{3}H, the consequence can be a deviation of detection efficiencies calculated with the statistical TDCR model. This deviation is due to the stochastic time dependence between PMTs, which is not considered in the statistical TDCR model (Bobin et al., 2012a).

The evolution of double-coincidence counting rates according to increasing resolving times was compared for a 2.7 keV-energy emission using two liquid scintillators, Hionic Fluor (HF) and Ultima Gold (UG). For these measurements, the x-ray beam was not collimated in order to have interactions through the whole surface of the liquid scintillator. The experimental results given by the time-to-digital converter especially designed for TDCR measurements are plotted in Fig. 2, taking the counting rates for a coincidence resolving time set at 40 ns as a reference (~220 s^{-1} for HF; ~400 s^{-1} for UG). Similarly to the results previously obtained with radioactive LS sources, double-coincidence counting rates are sensitive to increasing resolving times (especially below 120 ns for HF and 200 ns for UG). The difference of behavior between the two scintillators is in agreement with the hypothesis proposed in Bobin et al. (2012a), that the PMT–PMT coincidence timing distribution depends on the scintillation lifetime of the liquid scintillator used. The higher sensitivity observed for UG can be explained by the fact that the solvent is a naphthalene derivative molecule (di-isopropylnaphthalene) which is known to generate triplet–triplet annihilation that yields delayed fluorescence applied for alpha/beta discrimination (Aupiais et al., 2003).

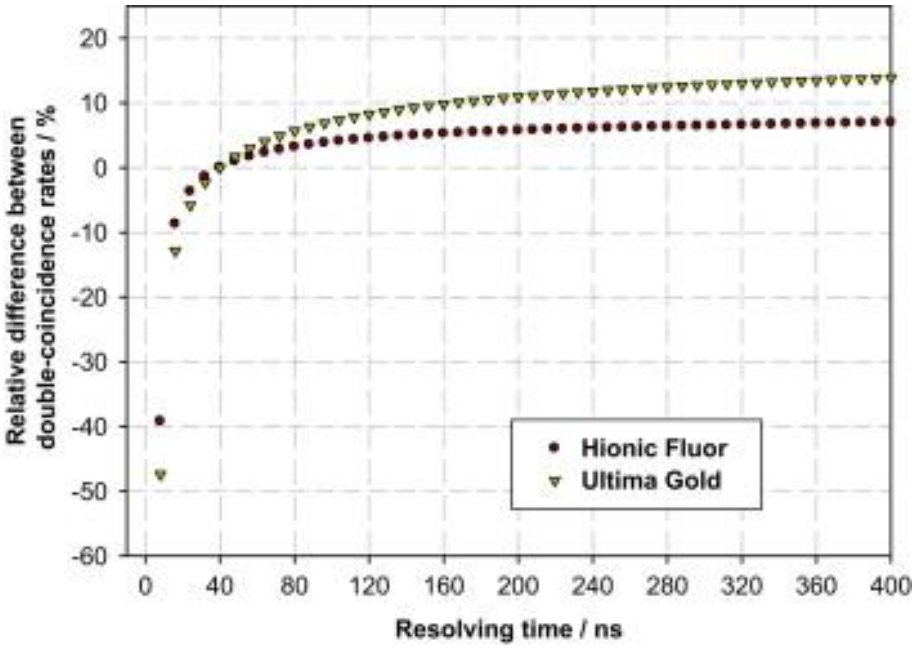

Fig. 2. Comparison between two liquid scintillators (Hionic Fluor and Ultima Gold) of the evolution of double-coincidence counting rates with increasing resolving times for an energy deposition of 2.7 keV (the collimator was not used). The relative differences are calculated using a reference value given by counting rates corresponding to a resolving time of 40 ns.

Additional measurements carried out with the UG scintillator have shown that the type of vial (glass or polyethylene) has no significant impact on counting rates with regard to the influence of coincidence resolving times. On the other hand, as displayed in Fig. 3, a significant shift is observed for the relation between the double coincidence rates and the experimental *TDCR* value. As previously studied with the TDCR-Geant4 model, this effect reflects the influence of the geometrical stochastic dependence between PMTs (i.e. due to geometrical optics) on triple and double coincidence counting (Bobin et al., 2012a).

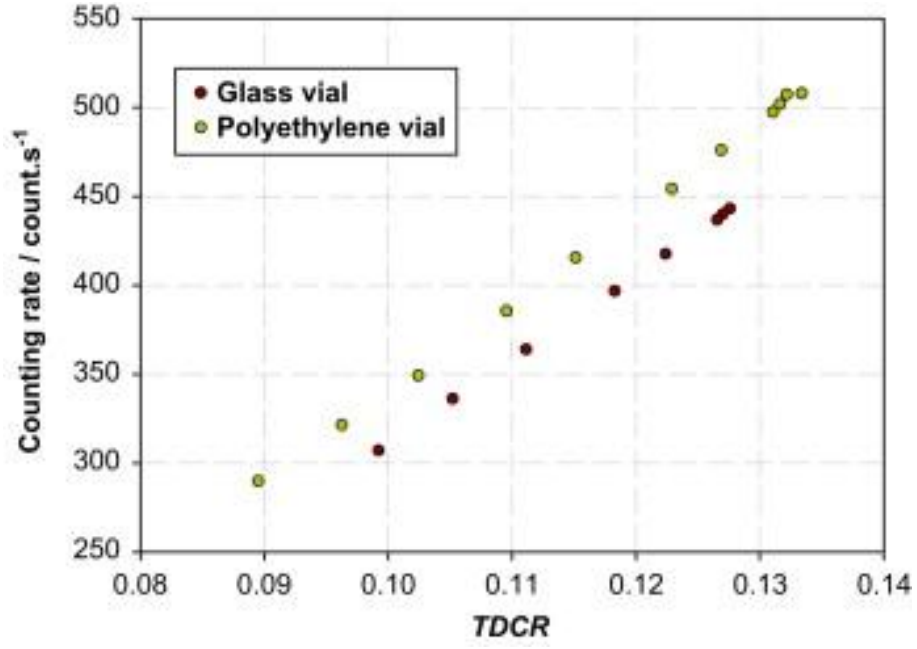

Fig. 3. Comparison between polyethylene and glass vials of double-coincidence counting rates versus *TDCR* values for x-ray photons of 2.7 keV (the collimator was not used).

4. TDCR-Geant4 modeling

The Monte Carlo Geant4 code was chosen for its capability to simulate the transport of ionizing and electromagnetic radiations (Agostinelli et al., 2003). The simulation results obtained in the present study were performed with the Geant4.9.4 version using the low-energy electromagnetic physics based on PENELOPE models (Apostolakis et al., 1999). The model also includes the atomic relaxation using the Livermore Evaluation Atomic Data Library, which contains the radiative and non-radiative transition probabilities for each sub-shell of each element, for Z=1–100 (Perkins et al., 1991). The Geant4 package allows for the transport of optical photons using the UNIFIED model developed for the DETECT project (Levin and Moisan, 1996). For that purpose, the geometrical model of the LS counter is constructed with the optical properties (according to the photon wavelengths) attached to each element of the optical cavity. Optical photons are simulated taking into account refraction and reflection processes (depending on refractive indexes of medium boundaries) and spectral transmittance of materials (borosilicate vial, PMT windows).

As already described in previous studies, the TDCR-Geant4 modeling has been first investigated with the application of the TDCR-Cherenkov technique (Bobin et al., 2010b, Thiam et al., 2011). The TDCR-Geant4 benchmark is able to simulate the transport of ionizing particles emitted in aqueous solutions and subsequent Cherenkov photons. Optical photons are tracked from their creation (considering the anisotropy of Cherenkov emission) inside the optical chamber (aqueous solution, glass vial, PMT windows) to their conversion into photoelectrons at the PMT photocathode. For each energy deposition following a disintegration, a binomial trial is applied to all the photoelectrons produced in the PMTs in order to determine if they are detected or not, and then to calculate double and triple coincidences between PMTs. This trial represents the probability for a photoelectron to reach the first PMT dynode and it is used to simulate the PMT defocusing for the detection efficiency variation. The probability to count at least one photoelectron in a PMT calculated with the TDCR-Geant4 model differs from the statistical modeling by taking into account potential stochastic dependence between PMTs. Activity measurements based on Cherenkov emission were carried out for various radionuclides: ^{90}Y, ^{11}C, and ^{32}P (Bobin et al., 2010b, Thiam et al., 2011). Because of the anisotropy of Cherenkov emission, these results are considered as a validation step of the geometrical modeling. As for the classical statistical model, the extension of the TDCR-Geant4 model to scintillation counting is based on the Birks formula with regard to the non-linearity of light emission as a consequence of ionization quenching. In that case, the TDCR-Geant4 model has been applied to activity measurements of ^{63}Ni, ^{51}Cr and ^{60}Co in UG (Bobin et al., 2012b, Thiam et al., 2012).

5. Estimation of the *kB* factor and scintillation yield used in the Birks formula

When the statistical TDCR method is implemented, the activity is usually determined from the experimental *TDCR* values obtained by variation of the detection efficiency. An optimal *kB* factor is estimated in order to obtain the most consistent activities over a range of experimental *TDCR* values. As it is included in a free parameter, the value of the scintillation yield (characterizing the liquid scintillator) is not explicitly specified for the activity determination (Broda et al., 2007). In the case of TDCR-Geant4 model, the scintillation process is also implemented using the Birks formula but the scintillation yield has to be defined in terms of photons emitted per keV. The measurement of this parameter was previously carried out using the 4π(LS)β−γ coincidence technique (Bobin et al., 2012a).

For a given liquid scintillator and x-ray energy, pairs of *kB* factor and scintillation yield are calculated with the TDCR-Geant4 model using the TDCR measurement obtained with the x-ray tube coupled to the LS counter. For the preliminary tests of this system, the measurements were carried out using a glass vial containing 10 mL of UG. The size of the x-ray beam was collimated using a Delrin collimator (*Ø*=8 mm) in order to decrease the diameter of the volume where x-ray photons interact in the liquid scintillator. In this configuration, the geometrical stochastic dependence on TDCR measurements resulting from the photon distribution between PMTs is reduced (Bobin et al., 2012a). The aim was also to minimize possible biases due to the transport of particles and photons in the meniscus region which is considered in the Geant4 modeling (Thiam et al., 2011). The experimental *TDCR* values obtained for x-ray energies equal to 2.7 keV, 8.7 keV and 17.3 keV are respectively 0.137 (2), 0.646 (2) and 0.928 (2). As expected with the TDCR-Geant4 model, the collimation increases the measured *TDCR* values: for instance, at 2.7 keV *TDCR*=0.128 (2) without the collimator.

In the TDCR-Geant4 modeling, the x-ray emission is implemented according to a beam flux with energies randomly generated using the spectra measured with the SDD for each energy. The spectrum obtained in the case of 2.7 keV x-ray photons is displayed in Fig. 4. Simulations were carried out for 2.7 keV photons according to different configurations of energy distribution: mono-energy, monoenergy convoluted with a Gaussian function (using the energy resolution of the SDD) and the x-ray spectrum measured with the SDD. The results have shown no significant differences within uncertainties. The atomic rearrangement resulting from x-ray interactions with the main components of the UG scintillator (~79% carbon, ~10% hydrogen, ~9% oxygen and ~1.4% phosphorus) is also considered in the Geant4 simulations. The simulated photoelectron spectrum given by 2.7 keV x-ray interactions in UG is shown in Fig. 5. Most of the energy distribution is comprised between 2 keV and 2.7 keV (~85%). The photoelectron spectrum also features a non-negligible component at about 600 eV (~12%) due to the photoelectric effect with phosphorus in the liquid scintillator.

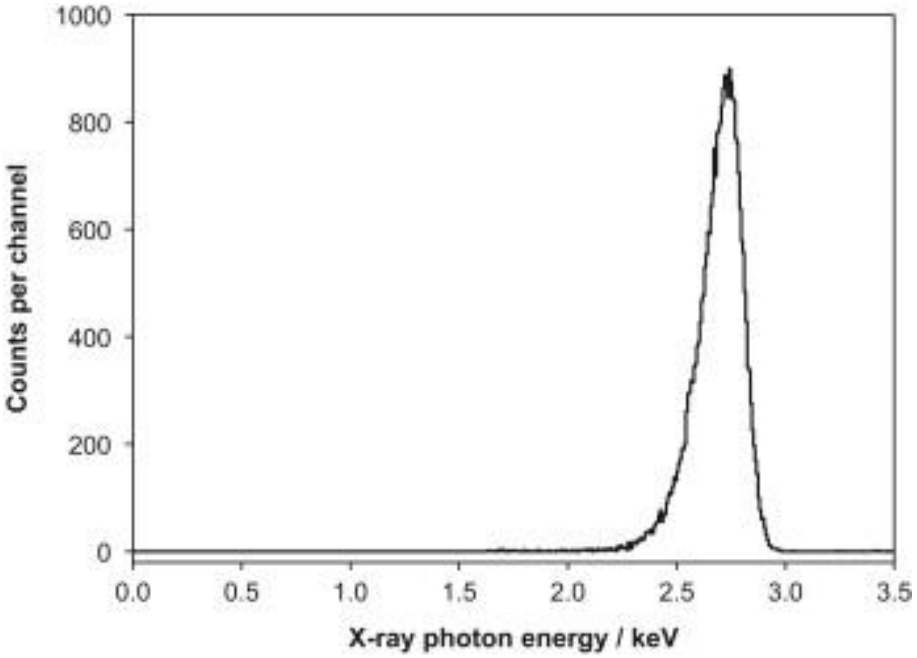

Fig. 4. Energy spectrum obtained with a SDD of x-ray photons emitted by the miniature x-ray tube tuned to 2.7 keV. The energy resolution is mainly due to the detector.

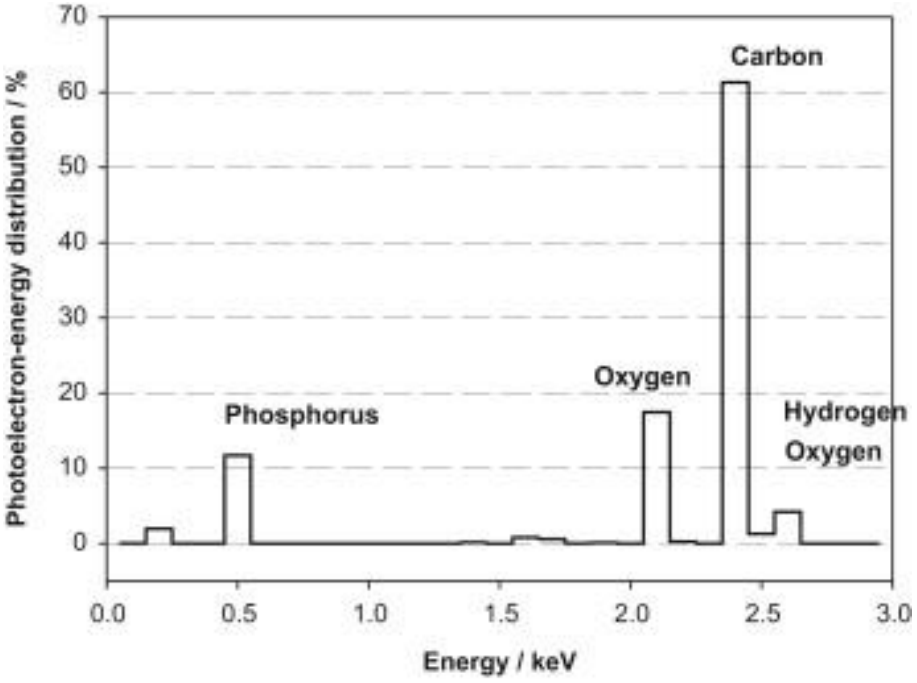

Fig. 5. Simulated photoelectron spectrum resulting from 2.7 keV x-ray interactions in UG.

For each photon energy obtained with the x-ray tube, several simulations with the TDCR-Geant4 model were carried out for *kB* factors ranging from 0.07 to 0.13 mm MeV^{-1} (corresponding to usual values drawn from the literature) and scintillation yields (ranging from 7 to 10 photons keV^{-1}). The pairs of *kB* factor and scintillation yield displayed in Fig. 6 were obtained by minimizing the difference between measured and calculated *TDCR* values (e.g. in the case of the 2.7 keV x-ray energy, each pair of *kB* factor and scintillation yield corresponds to a *TDCR* value equal to 0.137 (2)). In the range of *kB* factors and scintillation

yields investigated, it appears that the plots follow a straight line with a slope which decreases when x-ray energy increases. Considering that the pair of *kB* factor and scintillation yield is constant with energy in the Birks formula, a coherent pair can be obtained: kB=0.13 mm MeV^{-1} associated with a scintillation yield equal to 8.9 (1) photons keV^{-1}. In future, the range of the *kB* factor will be extended above 0.13 mm MeV^{-1} for further simulations.

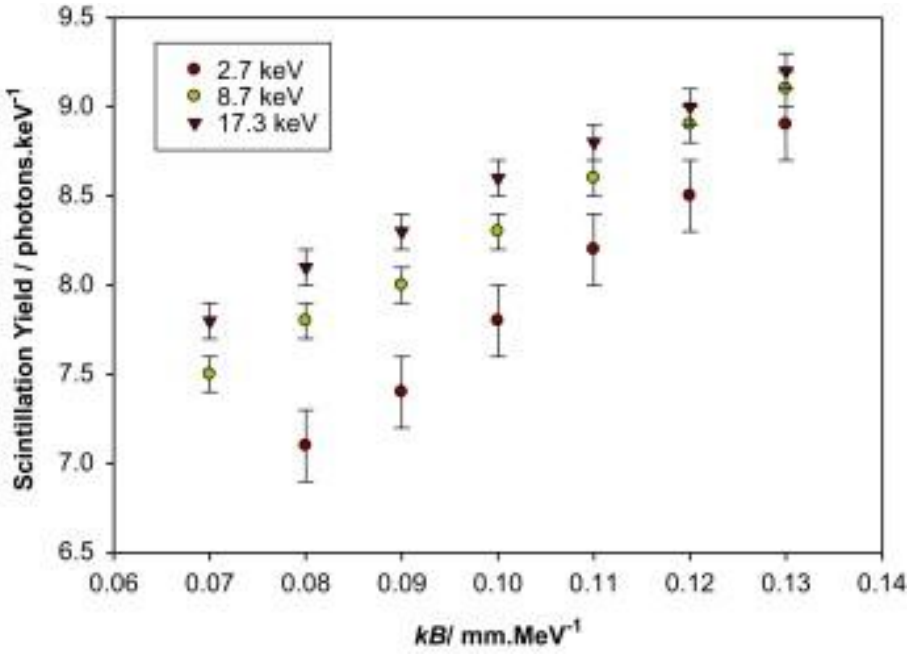

Fig. 6. Evolution of [*kB* factor; scintillation yield] pairs for three different energies obtained with the miniature x-ray tube.

Based on detection efficiencies measured with 4π(LS)β−γ coincidence counting (Bobin et al., 2012a), a [*kB*; scintillation yield] pair equal to [0.1 mm MeV^{-1}; 8.2 (3) photons keV^{-1}] in the liquid scintillator UG was previously assessed from ^{54}Mn measurements (photon energy mainly comprised between 5 and 6 keV). This first result is in agreement with those reported in this work for 8.7 keV photons given by the x-ray generator: [0.1 mm MeV^{-1}; 8.3 (1) photons keV^{-1}] with the same detection set-up.

6. Application of the TDCR-Geant4 model to the standardization of ^{3}H

The results obtained with the x-ray generator [kB=0.13 mm MeV^{-1}; 8.9 (1) photons keV^{-1}] were used in the TDCR-Geant4 model for activity measurements of ^{3}H (β-emitter; E_{max}=18.6 keV) in UG. Activity concentrations were calculated from measurements obtained by PMT defocusing (*TDCR* values ranging from 0.49 to 0.54) using both the TDCR-Geant4 and statistical models. As observed in Fig. 7, the activity concentrations are about 1.3% higher in the case of the TDCR-Geant4. Moreover, the slope featuring activity concentration as a function of *TDCR* values is lower for the results given by the statistical model. Additional plots in Fig. 7 were obtained with the TDCR-Geant4 model by limiting the electron emission at the center of the scintillator volume: the slope difference between both models vanishes. Because this configuration of Geant4 simulations attenuates stochastic dependence due to geometrical optics, it is considered to be closer to the statistical model (Bobin et al., 2012a).

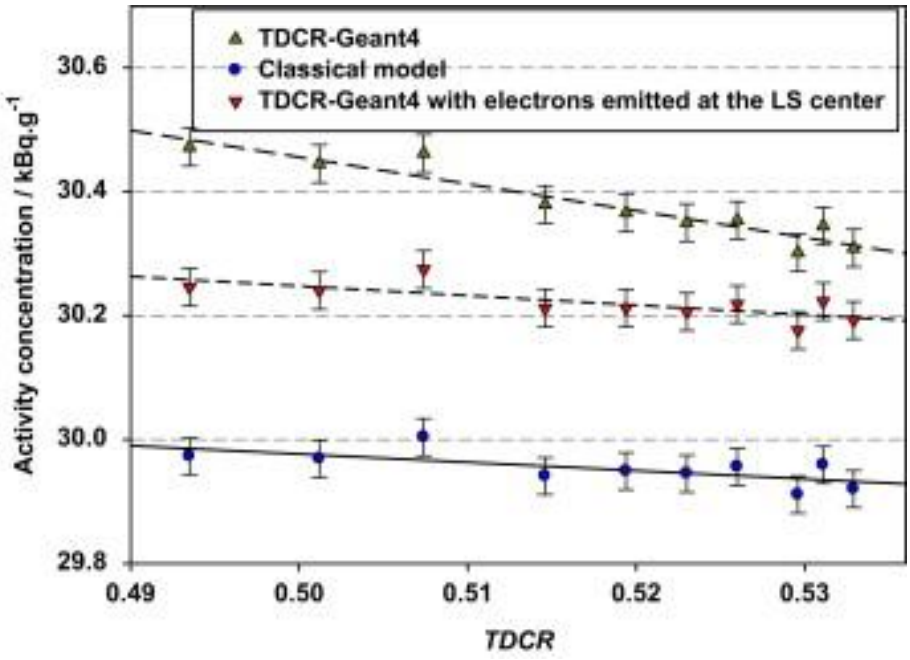

Fig. 7. Activity concentration versus *TDCR* value in the case of ^{3}H in UG obtained with both statistical and TDCR-Geant4 models (*kB* equal to 0.13 mm MeV^{-1}).

Due to the low energies of emitted electrons, the standardization of this β-emitters such as ^{3}H is sensitive to the *kB* factor. In that case, the *kB* factor is generally used as a second free parameter for the activity calculation with the statistical TDCR model. The usual practice is to optimize the *kB* value by minimizing the fitting slope of the function giving the activity concentration versus *TDCR*. Flat slope is considered as an indicator of the goodness of the model. For the present study, the optimal *kB* factor given by the statistical model is equal to 0.10 (1) mm MeV^{-1} leading to an activity concentration equal to 29.5 (2) kBq/g (the activity concentration corresponding to *kB*=0.13 mm MeV^{-1} is equal to 29.95 (20) kBq/g). In the case of the TDCR-Geant4, the slope (activity concentration versus *TDCR*) is less sensitive to the variation of the *kB* factor. As a result, the activity determination performed by minimization of the *kB* factor is not applicable with the TDCR-Geant4 model. This effect was already observed in the case of the standardization of ^{63}Ni (β^--emitter; E_{max}=66.98 keV) but the activity concentrations given by both Geant4 and classical models were in good agreement (Thiam et al., 2012).

Further studies are needed to understand the differences observed between both TDCR-Geant4 and statistical models for the standardization of ^{3}H. In particular, the influence of the stochastic dependence due to geometrical optics on the *kB*-optimization procedure will be investigated.

7. Discussion and perspectives

First measurements performed with an x-ray generator coupled with a TDCR detection set-up were presented. The first aim of this new system was the possibility to reproduce with low-energy x-ray photons (less than 20 keV) the observations related to stochastic dependence between PMTs previously obtained with radioactive LS sources. Resulting from geometrical optics, the influence of the vial type (glass and polyethylene) observed by a shift of the relation between double-coincidence rates versus *TDCR* values was confirmed with an x-ray

beam tuned at 2.7 keV (liquid scintillator: Ultima Gold). Due to the sensitivity of PMT–PMT coincidence timing distribution to scintillation lifetime, a difference in the evolution of double-coincidence rate with coincidence resolving time was also presented when comparing two commercial liquid scintillators (Ultima Gold and Hionic Fluor). These results have shown that the effect due to stochastic dependence between PMTs first identified with interactions in the whole LS volume can be also observed with x-ray interactions taking place in a smaller volume close to the surface of the liquid scintillator (50% of 2.7 keV x-ray photons are absorbed in a thickness of about 70 μm). Thus, the new experimental TDCR set-up including the x-ray tube can be used to test or to refine the TDCR-Geant4 model for activity measurements of low-energy emitters.

A technique using the x-ray tube has been tested to assess the pair of *kB* factor and scintillation yield used in the Birks formula for a given liquid scintillator. Calculations were based on the minimization of the difference between an experimental *TDCR* value obtained for a given x-ray energy and calculations performed with the TDCR-Geant4. From measurements obtained with three x-ray energies (2.7 keV, 8.7 keV and 17.3 keV), a preliminary estimation of a pair of *kB* factor and scintillation yield is proposed: [0.13 mm MeV^{-1}; 8.9 (1) photons keV^{-1}]. This estimated *kB* factor was higher compared with the value given by the statistical model (*kB*=0.10 mm MeV^{-1}) obtained by minimizing the fitting slope of the function of the ^{3}H activity concentration versus *TDCR* values. The origin of the higher *kB* factor given by the TDCR-Geant4 model needs to be investigated. As previously observed for the standardization of ^{63}Ni, the slope (activity concentration versus *TDCR*) is not as sensitive to the variation of the *kB* factor as for the statistical model. As a result, the activity calculation with the TDCR-Geant4 model cannot be implemented by minimizing the slope with the variation of the *kB* factor. This result could be due to the fact that the TDCR-Geant4 model includes geometrical and optical properties of the LS counter. Finally, further investigations are planned to understand the difference between both TDCR models on activity measurement of ^{3}H by using other scintillation cocktails (e.g. containing no phosphorus) or by measuring other low-energy emitters (^{55}Fe, ^{241}Pu).

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